

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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IN THE APPLICATION OF: DAVID HOBSON

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TITLE: High Solids Content Dispersions

Wickliffe, Ohio

Dated: 8 June 2009

Hon. Commissioner for Patents  
P. O. Box 1450  
Alexandria, VA 22313-1450

Declaration Under Rule 132

Sir,

I, Claire Hollingshurst, declare as follows:

I received a Masters of Chemistry degree from York University in August 2000.

I have been employed by The Lubrizol Corporation since August 2000. From August 2000 to March 2007 I was a chemist in Chemical Synthesis responsible for the development of dispersants and detergents for the corporation. In March 2007 I was promoted to Team Supervisor for the Applied Sciences Dispersions Group. Within this role I am responsible for development of high solids content dispersions.

I have read and understand the subject matter of US Patent Application with Serial Number 10/598,577.

I have read and understand the subject matter of Forsberg (US 4,094,801), Crawford (EP 0 288 296), Young (GB 1 061 161) and Magyar (US 5,851,961).

After reviewing Forsberg, a comparative study was undertaken to evaluate the compositions of Forsberg against the high solids dispersions of the present invention. The relative amounts of solids, surfactant, oil content and water content for the 16 examples of Forsberg are calculated as is shown in the table below.

	Amount of Ingredient Added by Forsberg (parts)								
Ref	Solid	Surfactant	Solvent	Other	Oil	Water	% water	% solid	Product Form
1	135	600	0	0		50	6.37	17.20	firm gel
2	225	600	0	20		75	8.15	24.46	dark brown gel
3	225	600	900	48		75	2.73	8.19	soft brown gel
4	223	600	770			1250	7.93	14.74	soft gel
5	600	308	478		244	381	18.95	29.84	gel
6	320	204	515		88	420	9.62	25.66	gel
7	835	425	856	67	495	1106	29.23	22.07	gel
8	550	225	125		680	200	9.69	33.95	gel
9	745	294	165		895	2309	26.51	26.09	gel
10	333	132	75		401	900	6.37	33.13	opaque liquid
11	440	200	310		880	200	6.75	26.99	pourable gel
12	373	331	247		210	754	39.37	19.48	-
13	373	331	247		210	754	39.37	19.48	-
14	206	540				69	8.47	25.28	solid
15	552	180			505	1280	15.68	37.63	-
16	180	16		96	377		0.00	26.91	grease

The closest dispersion to that claimed in the present invention is Example 10. The reason is because it has the combination of the highest solids content with the least amount of water, and wherein the final product is a liquid.

The following examples and analysis were carried out under my supervision.

Forsberg Example 10 (COMPA)

The example of Forsberg was prepared by the following laboratory procedure. A mixture of 900 parts of water, 333 parts of magnesium oxide, 266 parts of mineral oil and 132 parts of the alkylbenzenesulfonic acid is heated to 80 °C, with stirring, over about 45 minutes. It is maintained at this temperature for about 15 minutes after gelation takes place.

Forsberg then discloses that a water layer forms and 793 parts of water was decanted followed by an addition of 135 parts of mineral oil and 75 parts of toluene, allowing an additional 43 parts of water to be removed by azeotropic distillation. Then the toluene was stripped at 180 °C by nitrogen blowing and the residue was screened to yield the desired magnesium oxide-sulfonate complex as a thick opaque liquid.

This did not occur so the following was attempted.

A further hold period of 1 hour 45 minutes was applied and then the reaction mixture was left to stand for 1 hour. There was still no gellation or separation after this timeframe. Stirring was restarted at this point and the mixture was held at 80 °C for a further 2 hours.

At this point, with no separation or gellation, the mixture was split in two.

The first half was charged to a 1L flask and 45 parts of toluene was added. The material was heated to 90 °C and held at this temperature for 2 hours to try and drive off the water into the Dean and Stark trap. Only 5 ml of water was collected so the reaction was terminated.

Since the preparative study did not gel as noted by Forsberg, a modified procedure was used to prepare the material of example 10. The procedure is shown below. The main difference in the procedure is that vacuum has been used to remove the toluene and water from the reaction. However, this change should not alter the product formed because the resultant material will be of the same composition and the temperatures used will not have affected the product.

The product was stripped using a Rotary evaporator initially set at 60 °C and low vacuum. The temperature and vacuum was increased until 90 °C and full vacuum (-28 in Hg equivalent to -711 mm Hg) was reached and all the available water had been removed. The product obtained was a thick opaque liquid that contained large solid particles. The final product contains about 6.4 wt % of water.

#### Example 10 Mixed by High Shear Stirring (COMPB)

The same process as COMPA, except the product is mixed using a Turrax™ rotastator mixer for 5 minutes to break up the large particles. Particles with a diameter of 2-3 mm did not pass through the mixer head and blocked the system, however the particle size was reduced.

#### Example 10 by Ball Milling (COMPC)

The same as COMPB, except the resultant material from the Turrax™ mixer was then charged to a vertical ball mill system, 500ml glass round-bottomed flask with 700g SLG 37/43 (Ø3.7-4.3 mm) glass beads and a U-shaped polypropylene stirring paddle. After 30 minutes of mixing the material had become very thick and stirring was impossible and had to be terminated.

Based on how viscous the product of COMPC had become during preparation, I believe that the only two ways to make a material useful would be to add additional

oil, or significantly reduce the amount of magnesium oxide. Either of these two options would result in the solid content of the product being reduced.

#### Invention Example

A high solids dispersion of the present invention was prepared to be substantially similar to example 10. The dispersion is prepared by taking a mixture of 990 parts magnesium oxide, 390 parts of the alkylbenzenesulfonic acid, and 1620 parts of mineral oil. The mixture was blended together for 30 minutes, until smooth and homogeneous, using a high shear, saw tooth stirrer. This slurry was then milled in 3 passes using an ECM Dyno-mill (WAB) with a 65% fill of Ø0.5 mm YTZ (Yttrium-zirconium) beads. The separation gap for the mill was set to 0.2 mm, the cooling water set to 10°C, and the tip speed set to 8 m/s. During the three passes the mill temperature was 55-65°C, and the flow rate was 0.162 L/min on average (the flow rate changes considerably as the milling progresses, this figure is an average flow rate for the three passes and was taken mid milling for each pass). The final dispersion is an opaque liquid.

#### Example Summary and Characterisation

The preparative example from example 10 of Forsberg, the milled dispersion of Forsberg and the dispersion of the present invention are summarised and characterised as described in the table below.

The viscometric data was obtained by using a TA Instruments AR2000™ rheometer using a flat plate geometry looking at viscosity versus shear rate performance.

The particle size data was obtained by using a Coulter® LS230 light scattering particle sizer.

	Forsberg COMPA	Turrax Mixer Forsberg COMPB	Ball Milled Forsberg COMPC	Invention
Solid Type	Magnesium oxide	Magnesium oxide	Magnesium oxide	Magnesium oxide
Solid Content	33 wt%	33 wt%	33 wt%	33 wt%
Surfactant	Alkylbenzene sulfonic acid	Alkylbenzene sulfonic acid	Alkylbenzene sulfonic acid	Alkylbenzene sulfonic acid
Surfactant Content	13 wt%	13 wt%	13 wt%	13 wt%
Oil Content	47.6 wt%	47.6 wt%	47.6 wt%	54 wt%
Water Content	6.4 wt%	6.4 wt%	6.4 wt%	0 wt%
Physical State	Thick Opaque Liquid	Thick Opaque Liquid	Thick Opaque Liquid	Opaque Liquid
Viscometrics				
Shear Rate 0 1/s	2771 Pa.s	321 Pa.s	2477 Pa.s	12.99 Pa.s
Shear Rate 500 1/s	1.759 Pa.s	2.1 Pa.s	1.5 Pa.s	1.195 Pa.s
Particle Size characterisation (Mean $\mu\text{m}$ )	60.62	31.16	10.27	0.937

From the data obtained above, it is evident that the Forsberg material is much more viscous on standing (i.e., no shear), and contains much larger particles than the invention dispersion. Upon increasing the shear rate to 500 1/s, the viscosity of all the comparative examples and the invention example are approximately similar, to the extent that the difference in viscosity will have negligible effect on performance.

#### Test: Stability in Diesel Fuel

The samples were then evaluated by the following test procedure. The samples were diluted into 40 g of middle distillate (diesel fuel) at the following treat rates: 1000, 1500, 2000, 2400, and 3000 ppm. Once the samples had been added to the diesel at the required treat rate the mixtures were shaken until the sample had fully dissolved and then left to stand to be rated for sedimentation.

The test evaluates performance of fuel additives in diesel fuels for storage stability. The results obtained were:

Treat rate to diesel (ppm)	Forsberg COMPA		Forsberg COMPB		Forsberg COMPC		Invention	
	Stability	Rating	Stability	Rating	Stability	Rating	Stability	Rating
1000	<1	F	<1	F	<1	F	5 to 6	A
1500	<1	F	<1	F	<1	F	5 to 6	A
2000	<1	F	<1	F	<1	F	5 to 6	A
2400	<1	F	<1	F	<1	F	5 to 6	A
3000	<1	F	<1	F	<1	F	5 to 6	A

Footnote:

Stability units is measured in hours

<1 – indicates that once the sample had fully dissolved and was then left to stand sedimentation occurred soon afterwards due to the large particle size of the Forsberg product.

5 to 6 – indicates that the sample was stable after 5 hours, but upon inspection at 6 hours showed signs of sedimentation

A = Top 7% transparent, light easily re-dispersible solid

F = Almost 100% transparent, heavy sediment

#### Discussion and Conclusion

The results indicate that the dispersion of the invention was stable in the fuel for a period of 5 to 6 hours. In contrast the product of Forsberg (COMPA) is unstable after less than one hour. The same stability, less than 1 hour, was also observed for both the Turrax™-blended sample COMPB and the ball milled Forsberg sample (COMPC). Consequently, the dispersion of the present invention has improved storage stability in the field. This means that when the fuel is used benefits may include reduced sedimentation, improved transportation/pumpability with fewer blockages. Smaller particle sizes would lead to improved performance in terms of reactivity also. These benefits would not be realised to the same extent by the Forsberg dispersion because this dispersion is unstable when treated to fuel so may form blockages in the pipelines or delivery system, and also reduce the actives levels being delivered due to sedimentation prior to delivery.

I further declare that all statements herein made of my own knowledge are true and all statements herein made on information and belief are believed to be true. I understand that wilful false statements and the like are punishable by fine or

imprisonment or both (18 U.S.C. 1001) and may jeopardize the validity of the application or any patent issuing thereon.

C. Hollingshurst  
Claire Hollingshurst

8<sup>th</sup> June 2009  
Date